X-ray Science at DESY: Upgrade programs for the user facilities FLASH and PETRA III

K. Baev, K. Bagschik, R. Bartolini, M. Gühr, S. Klumpp, A. R. Maier, A. Martinez de la Ossa, J. Osterhoff, H. Reichert, L. Schaper, S. Schreiber, C.G. Schroer, K. Schubert, P. Winkler, W. Leemans, E. Weckert

Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany

Introduction

The Deutsches Elektronen-Synchrotron (DESY) in Hamburg (Germany) is one of the leading particle accelerator centers. With more than 60 years of experience in the design, construction, and operation of large accelerator facilities, initially for particle physics, the current focus has shifted to photon generation using synchrotron radiation and free-electron lasers. Current machines in operation are the high-energy synchrotron radiation source PETRA III, the worldwide first VUV and soft X-ray free-electron laser FLASH, and the more than 1.7 km long superconducting linear accelerator of the European X-ray free-electron laser (European XFEL). As part of the Helmholtz Association of German Research Centers (HGF), one of DESY's missions is research in processes and technologies for a sustainable future and a competitive economy using accelerator-based light sources to decode the properties of matter.

DESY currently operates PETRA III, one of the world's brightest 3rd generation storage-ring-based X-ray sources for high-energy photons. It provides the environment for specialized experiments to address many of the pressing questions of the 21st century in the fields of energy, life science and health, earth and environment, mobility and information technology. With a circumference of 2304 m, it is the world's largest storage-ring-based source, operating at an electron energy of 6 GeV. PETRA III offers experimental techniques using mainly hard and high-energy X-ray radiation on currently 25 beamlines located in three separate experimental halls. The beamlines are optimized for different applications using nano-focused or highly collimated X-ray beams. Three of the beamlines are operated by the European Molecular Biology Laboratory (EMBL). Two other beamlines for materials science are operated by the Helmholtz Centre Hereon. Following the successful upgrade of ESRF-EBS and the running programs in US, China, Japan and Europe, DESY is preparing the project for the upgrade of PETRA III. The new facility, PETRA IV, will be based on an ultra-low emittance lattice, additional experimental stations and new access models. PETRA IV will put back DESY in a world-leading position with the highest X-ray photons brilliance source.

PETRA III is complementary to the soft X-ray user facility BESSY II operated by the Helmholtz Centre Berlin (HZB) [1]. The upgrade of the user facilities to PETRA IV and to BESSY III, respectively, would provide the German scientific and industrial user community with unique X-ray analytical tools from the infrared to the high-energy photon range.

The FLASH facility is a high-repetition rate VUV and soft X-ray FEL with the two undulator branches FLASH1 and FLASH2. The goal of the FLASH2020+ upgrade program is to increase the accessible spectral range, to provide highly stable near Fourier-limited as well as ultra-short pulses, and achieve fully independent operation of the two FEL lines. This will enable the user community to investigate dynamic processes in materials and chemical reactions on timescales that are not accessible today.

The DESY campus in Hamburg is embedded in the growing Science City Hamburg Bahrenfeld, a vibrant environment for academic and research institutions and deep-tech companies, including start-ups. The excellent research prospects at DESY are underlined by numerous national and international collaborations, e. g., with Universität Hamburg and Universität Kiel, the Max Planck Society, the Swedish Research Council and the Indian Jawaharlal Nehru Centre of Advanced Sciences. Many interdisciplinary research institutes on campus are using the X-ray photon user facilities, such as the Centre for Free-Electron Laser Science (CFEL), the Centre for Structural Systems Biology (CSSB), the Centre for X-ray Nanoscience (CXNS), or the EMBL, to name only a few.

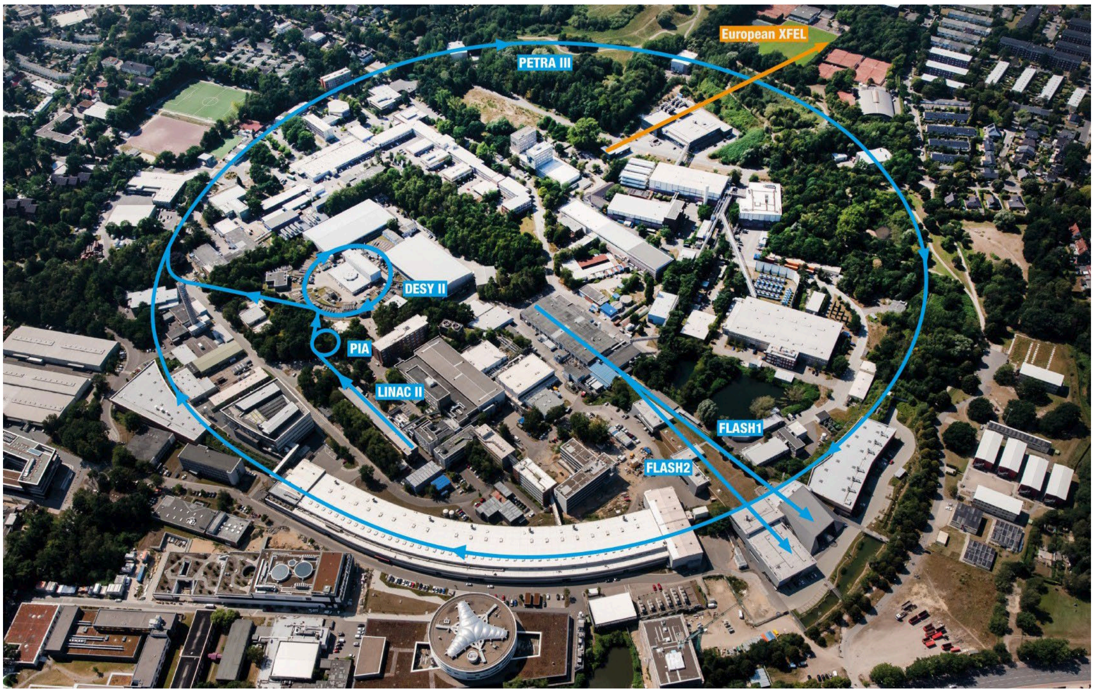

Figure 1: DESY campus in Hamburg with PETRA III, FLASH, and the first part of the linear accelerator of the European XFEL. (Credit: DESY)

PETRA IV – the ultralow emittance source for hard X-rays

Following the HGF's and DESY's mission statements, it is the goal of PETRA IV to provide analytical tools for science and innovation to decode the structure and function of matter down to the atomic scale. Building on the fundamental knowledge of structures and processes, new functional material components can be tailored. This facilitates the transition from the current, often empirical, approach to materials design to a knowledge-based, rational design of materials, drugs, and processes, enabling thus a new era of "observe and control".

The upgrade of PETRA III to PETRA IV will result in the world's brightest X-ray source for the next decades, enabling high-resolution 3D imaging of heterogeneous materials in real time, pushing spatial resolution towards the atomic scale, and allowing local quantitative measurements of physical and chemical properties from nanometric to macroscopic dimensions enabling efficient multi-scale imaging. Thanks to the deep penetration of hard X-rays, PETRA IV is ideal for studying samples in their natural environment and under working conditions (*in-situ/operando*). With a factor of about 500 increase in brilliance (cf. Fig. 2) at high photon energies as compared to PETRA III, the combination of the highest spatial resolution and sensitivity with the ability to measure physical and chemical properties simultaneously *in-vivo*, *in-situ*, and *operando*, PETRA IV will be the ultimate 3D X-ray microscope for biological, chemical and physical processes. X-ray analytical studies can be carried out

- up to 500 faster. It will be possible to record movies of processes with highest spatial and temporal resolution and with chemical, structural, magnetic, and electronic contrast,
- with up to 25 higher sensitivity. The sensitivity of X-ray analytical techniques can be increased by more than an order of magnitude compared as to today, enabling the detection of trace elements and nanoscopic features that are undetectable today, and
- with up to 500 larger field of view or sample volume, allowing to image large objects with microscopic detail or analyze a large number of samples.

Scope of the Upgrade Project

The multi-bend achromat technology for the next generation of ultra-low-emittance synchrotron radiation sources combined with the large circumference of PETRA offers the unique opportunity to push synchrotron radiation close to the physical limits at around 5 keV photon energy. In order to make optimum use of the new capabilities, DESY plans to operate PETRA IV within a new business model that comprises improved, flexible, and rapid access for all users and extended support in particular for new and unexperienced users. Applications for access can be submitted at any time. After review and ranking by external experts, access should be scheduled within a few weeks, if requested in the proposal. Access to any of the complementing analytical infrastructures of PETRA IV can be applied for with or without beamtime.

The PETRA IV project comprises the upgrade of the present synchrotron radiation source PETRA III to an ultra-low-emittance source. The main goal is to replace the PETRA III storage ring with a state-of-the-art ultra-low-emittance storage ring based on hybrid multi-bend achromat (HMBA) technology. This upgrade includes the renewal of parts of the existing pre-accelerator chain and of the storage ring infrastructure. The new storage ring can store relativistic electrons in a beam that is 50 to 100 times more focused and less divergent than currently possible at PETRA III. By generating synchrotron radiation in specialized new undulators, photon beams can be obtained with a spectral brilliance 60 to 830 times higher than available today (see Fig. 3), outperforming all current and planned synchrotron radiation sources worldwide.

This outstanding performance of PETRA IV can only be exploited with a new generation of experimental stations occupying their own dedicated sector along the storage ring. At PETRA III, most straight sections are canted for hosting two beamlines in the same sector.

Figure 2: PETRA IV complex (buildings marked in dark blue): besides several technical buildings, it comprises the three existing experimental halls and a new experimental hall in the western part of the DESY campus. (Credit: DESY)

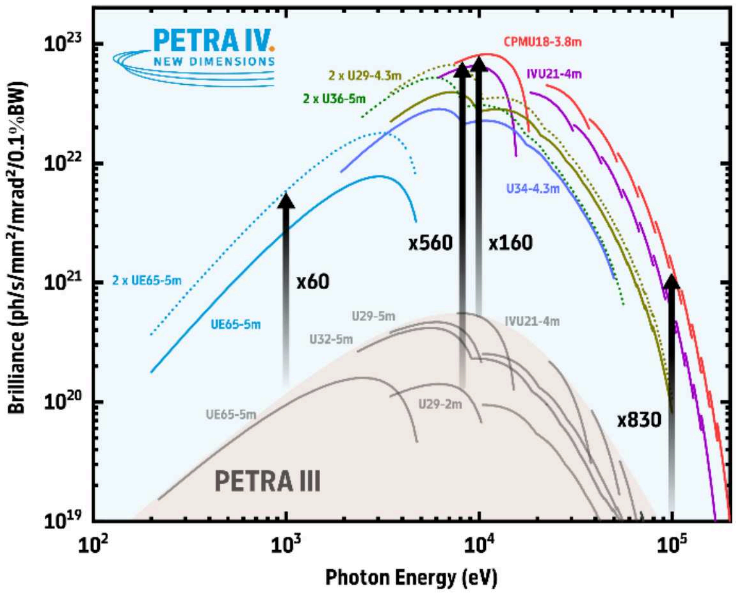

Figure 3: Comparison of spectral brilliance of PETRA IV (H6BA lattice) and PETRA III. The different colors denote undulator design optimized for different photon energy ranges. Vertical arrows denote the gain in brilliance. (Credit: DESY)

At PETRA IV, each beamline will occupy its own sector (with some exceptions), which requires to move some beamlines to new sectors. The project includes the construction of a new, large, experimental hall in the western part of the DESY campus providing 18 sectors for beamlines. Besides accommodating some of the moving beamlines, this new experimental hall also provides capacity for new beamlines and complementary analytical facilities satisfying future scientific and industrial needs.

X-ray microscopy techniques in particular will be advanced in a transformative way at PETRA IV:

Coherent X-ray microscopy: Coherent X-ray diffraction microscopy (e. g., ptychography or Bragg coherent X-ray diffraction) and holographic techniques gain in resolution, field-of-view, and sensitivity as brilliance increases. Imaging experiments taking weeks or months nowadays will be feasible within days, e.g. the large-scale scanning of entire microchips. This will open up new opportunities for quality management, e.g. in industry.

High-energy X-ray techniques: The spectral brilliance of PETRA IV at 100 keV will be higher than that of PETRA III at its optimal energy of about 5 keV, and almost three orders of magnitude higher at the same energy. This enables efficient high-energy X-ray nano-focusing that is currently out of reach. Techniques such as X-ray microscopy with Compton scattering contrast, high-resolution diffraction contrast, or with local structural contrast using the pair-distribution-function (PDF) technique are benefitting tremendously from this increase in performance.

While Compton scattering contrast has particular applications in biological imaging, high-resolution X-ray scattering opens new avenues for revealing structural details in complex engineering materials. For example, tomographic pair distribution function mapping will provide access to the local 3D structure of disordered complex materials such as catalysts, batteries, microelectronic and micromechanical systems, nanocomposites, biomaterials and medical implants. With nano-focused high-energy X-rays, the dynamics of engineering materials can be followed *in-situ* on the nanoscale, during heat treatment, plastic deformation, or phase transitions to name but a few.

PETRA IV Storage Ring

The current design of the PETRA IV storage ring is based on a Hybrid 6-Bend Achromat (H6BA) lattice (see Fig. 4) [2]. This design combines optimal performance in terms of machine parameters and beam stability (see Tab. 1), as well as compatibility with the current arrangement of beamlines on the PETRA III experimental floor. The natural emittance of the H6BA lattice (45 pm·rad) is reduced to the target emittance of 20 pm·rad by the use of additional damping undulators. This has the added benefit of providing efficient and reliable emittance control. The bare H6BA lattice features a remarkably large dynamic aperture and momentum acceptance compared to the previous lattice designs [3,4]. The result is a storage ring with a large Touschek lifetime and stability, allowing operation with off-axis injection and accumulation, which could not be guaranteed with the previous lattice design. The target parameters for the H6BA lattice of PETRA IV are summarized in Tab. 1 and compared to the current parameters of PETRA III.

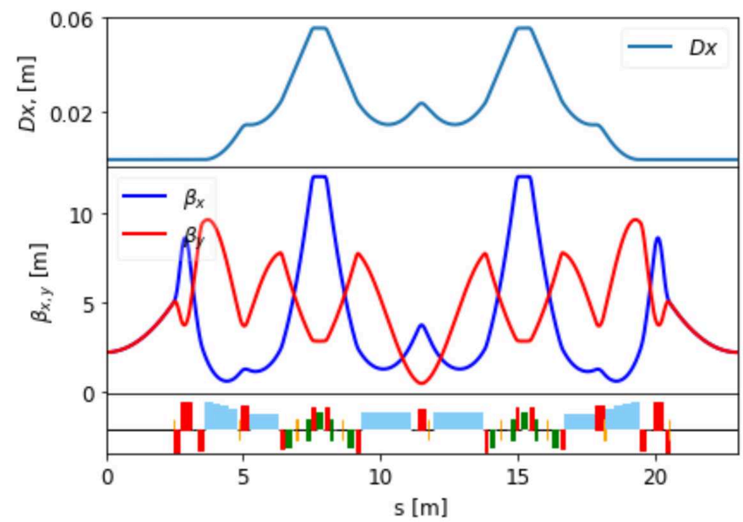

Figure 4: Hybrid 6-bend achromat cell of the PETRA IV reference lattice. (Credit: DESY)

Parameter	PETRA IV		PETRA III	
Mode	brightness	timing	continuous	timing
Storage ring energy	6 GeV		6 GeV	
Storage ring circumference	2304 m		2304 m	
Number of bunches	1600-1920	80 (40)	480 - 960	40
Total current / mA	200	80 (80)	120	100
Bunch current / mA	0.125	1.0 (2.0)	0.25-0.125	2.5
Arc ID β_x/β_y / m	2.2/2.2		high β: 20.0 / 4.0	
Long ID β_x/β_y / m	4.0/4.0		high β: 1.4 / 4.0	
Emittance				
Horiz. ϵ_x / pm rad	20	35 (38)	1300	
Vert. ϵ_y / pm rad	5	7 (8)	10	
Bunch length σ_z / ps	30	65 (75)	40	43
Bunch separation / ns	4	96 (192)	16 - 8	192
Energy spread σ_p / 10^-3	0.9	1.2 (1.5)	1.3	1.3
Touschek lifetime τ / h	> 10	> 5	9 - 13	1.5
Number of beamlines	35 + 1 VUV		24 (26) + 1 VUV	

Table 1: PETRA IV parameters as compared to PETRA III. PETRA IV will feature two modes of operation, one optimized for highest brilliance and one enabling time-resolved experiments.

PETRA IV Plasma Injector

Laser-plasma acceleration (LPA) has seen a surge in development over the past few years. DESY's mission is to turn plasma acceleration into a technology ready to drive its future facilities. As part of this endeavor, PETRA IV will test the feasibility of a plasma-based 6 GeV accelerator for direct injection into the PETRA IV storage ring, that would bypass the RF-driven Linac and DESY booster synchrotron.

An LPA-driven injector could, mainly due to the overall smaller footprint, offer a significantly reduced energy consumption and operating costs and would contribute to an overall more sustainable operation of the large-scale research facility. The layout of a future plasma injector is illustrated in Fig. 5 below.

For deployment in a user facility environment, LPA electron beam quality, as well as availability and reliability of the whole system need to be further matured. Antipov et al. have proposed an active RF-dechirper [5] to bring the LPA energy spread and jitter to the sub-permille level required for injection. Significant improvements in reliability and stability are expected from the ongoing push towards higher repetition rate drive lasers, that allow to deploy active stabilization and feedback techniques also in the laser-plasma domain.

These developments are tightly embedded into a roadmap that will verify the readiness of core technologies, such as laser pulse guiding and active RF-dechirping, and cumulate in testing a fully functional injector beamline. The roadmap will allow a timely and informed decision to which depth plasma accelerator technology will be integrated into the future PETRA IV facility.

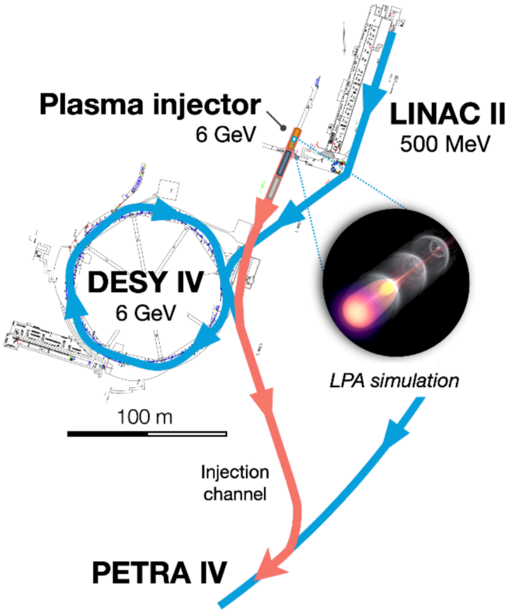

Figure 5 Schematic layout of a planned laser-plasma injector for PETRA IV operating parallel to the conventional pre-accelerator chain, which consists of a Linac and the booster synchrotron DESY IV. (Credit: DESY)

PETRA IV Beamlines

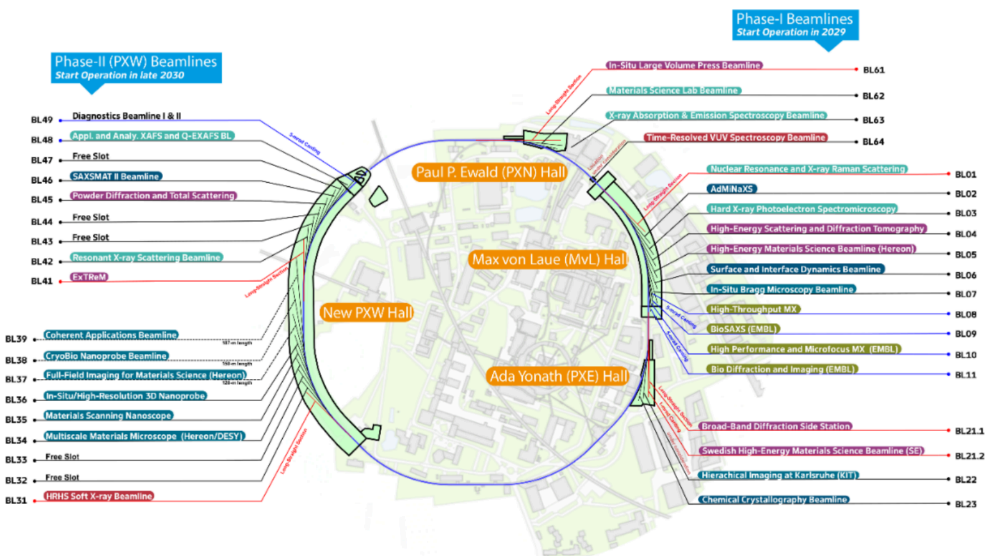

Figure 6: Beamline portfolio of PETRA IV and the beamline location in the four experimental halls. (Credit: DESY)

The beamline portfolio has been developed based on the scientific case, the unique beam properties of PETRA IV, and the needs of the user community at large. In early 2020, the global synchrotron radiation user community was invited to propose scientific instruments for PETRA IV. Workshops were held to foster collaboration and the sharing of experimental techniques. The resulting proposals were peer-reviewed, consolidated into beamlines by PETRA III and DESY experts, and refined with input from DESY's Photon Science Committee. The resulting beamline portfolio was subject to international review, focusing on PETRA IV science, user needs, productivity, technique diversity, and future upgrade possibilities.

An extensive screening of technology trend and foresight studies was undertaken to provide a comprehensive assessment of the beamline portfolio in terms of coverage of industry-relevant technology trends over the next 10-15 years [6]. The result confirmed that the beamline portfolio covers the needs of applied research and industry particularly well.

The beamline portfolio comprises 31 beamlines optimized for best performance as defined in the scientific and industrial case. It serves as the basis for the technical design and the funding proposal and will be updated and refined continuously throughout the detailed design and construction phase. Fig. 4 shows the distribution of the beamlines in the new facility. Within the next project phase, the conceptual design of all beamlines will be completed with a subsequent start of the technical design phase. 19 beamlines in the existing experimental halls should be operational in phase I shortly after the commissioning of the new storage ring. 12 more beamlines will then be built and commissioned in the new experimental hall in the West. Considering the dynamic development of economy, society and our environment, the remaining five free beamline slots provide sufficient flexibility to answer to new demands.

FLASH - The soft X-ray facility for ultrashort pulses and high stability

Soft X-ray Free Electron Lasers (FELs) enable the in-depth investigation of electron dynamics at unprecedented time resolution down to a few femtoseconds. While having operated two beamlines on the principle of self-amplified spontaneous emission [7], the FLASH2020+ project prepares the FLASH1 branch for externally seeded operation with high spectral and temporal stability and a level of longitudinal coherence close to the Fourier transform limit [8]. The project positions the source at a unique space in the landscape of FELs as the only externally seeded source with a high repetition rate.

The FLASH facility came into operation as the first FEL for users in 2005 [9]. Over the years the facility has been continuously improved, culminating in the construction of the second FEL beamline FLASH2, which showed first lasing in 2016 [10]. Employing a cryogenically cooled linear accelerator, the two FEL beamlines support up to 5000 freely distributable electron bunches per second and allow for two parallel user experiments at a time. The instrument portfolio at FLASH is highly tailored to its high repetition rate and allows unique experiments in various scientific areas. In condensed matter science, surface science, and photocatalysis on surfaces, X-ray photoelectron spectroscopy (XPS) is pursued successfully. The technique requires a high average flux and high stability due to space charge resolution limits. In AMO science, charged particle spectroscopy can be accomplished at dilute targets due to the high average flux combined with a high repetition rate. Time resolution in the femtosecond regime is either achieved via X-ray split-and-delay units or via pre-excitation of optical lasers. In addition, the high repetition rate of FLASH is ideal for pursuing X-ray emission and resonant inelastic X-ray scattering (RIXS).

For all the methods mentioned above, external seeding will be changing the acquisition speed and quality of the data, and thus attracting a wider user community. A prerequisite for the implementation of seeding was the improvement of the electron beam quality, tackled in a 9-month shutdown from November 2021 to August 2022, where a 70 m long section of the linear accelerator was renewed. Most notably, two of the superconducting modules have been replaced, boosting the beam energy by an additional 100 MeV to a maximum of 1.35 GeV. In addition, the two surrounding bunch compression sections with their respective diagnostics have been completely rebuilt. In the first bunch compression section, a laser heater has been added to reduce the gain factor of the so-called microbunching instability and, thus, allowing for reduced sliced energy spread at the radiator entrance. An increase in the SASE yield is observed in standard configuration and advanced operation modes with temporally tailored heating profiles have already been demonstrated to allow for control of the duration of the generated FEL pulses. The second bunch compressor has been replaced by a variable compressor, including skew quadrupoles, to act on transverse coupling effects while simultaneously optimizing the compression ratio. In addition, both compression sections also incorporate fast orbit correctors reducing transverse bunch offsets within a burst, and improved diagnostic sections. In the FLASH2 beamline, a so-called afterburner, an APPLE III type undulator boosting the third harmonic, was added to allow for user experiments with variable polarization at wavelength as short as 1.33 nm.

During the second phase of the upgrade from mid-2024 to mid-2025, the FLASH1 arm will be the focus. A completely new beamline tailored to external seeding will replace the existing infrastructure. Via high-gain and echo-enabled harmonic generation, a tunable seed laser, and APPLE III type radiators, FEL pulses at a spectral range from 60 nm down to 4 nm can be delivered to users with variable polarization. This provides unique benefits for XPS-based experiments, a spectrally stable source will allow for faster data acquisition and, thus, for systematic experiments in rapidly developing fields such as plasmonic photocatalysis [11]. For RIXS, the average spectral flux will increase even more due to the narrower bandwidth, which will have a direct impact on ultrafast phase-change studies in condensed matter and photochemistry. Apart from these improvements for already existing science areas, an externally seeded FLASH will also allow to explore new science areas. A powerful tool in chemical and biological research is ion traps, loaded by electrospray sources. The loading and read-out times of the devices do not allow for single-shot measurements. Full burst averaging at SASE sources will inevitably lead to washing out spectral features, and conversely a spectrally stable source will highly benefit these experiments. In addition, the external seeding at FLASH will boost schemes that benefit from the high repetition rate as well as from the high level of coherence. Stimulated RIXS will boost the applicability of the method due to the concentration of photons in a small detector area together with spectral selection [12]. Attosecond pulse trains from differently tuned seeded undulators pioneered at FERMI [13], will also be applicable to the high-repetition rate methods. To take full benefit, the FLASH user community is already starting to develop community proposals in different science areas. The process was initiated in a workshop in January 2024 and will continue up into the time-window between commissioning and regular operation with peer-reviewed experiments.

The surrounding infrastructure will also be prepared for future near-term upgrades. This enables fast implementation of a dedicated post-compression chicane to increase the efficiency of fully parallel THz generation and opens another opportunity for new experiments.

In the experimental halls the pump-probe lasers are part of the near-term upgrade strategy. Here, powerful main amplifiers and a modular concept directly at the beamlines, deliver increased pulse energy and repetition rate together with flexible wavelength conversion schemes covering a broad spectral range compatible with the multitude of experiments facilitated at FLASH. The upgrades culminate in a new beamline called FL11 opening up to the combined benefit of seeded beams together with THz and a pump probe laser at MHz repetition rate.

Shortest pulses providing highest time resolution for experiments have always been one of the driving factors in FEL science and experiments. Here, FLASH recently made significant progress by generating pulses as short as 1.2 fs [14]. For a potential mid-term upgrade renovating the FLASH2 beamline by the addition of a CEP stable laser to manipulate the electron bunch and a dedicated short undulator, simulations reveal that the generation of FEL pulses on the order of 100 attoseconds at nanojoule intensities is feasible. For longer time scales the Linac would be in the focus again, where severe modification to the accelerator and RF infrastructure would unlock quasi-continuous wave operation and, thus, increase the number of pulses available to users by 3 orders of magnitude.

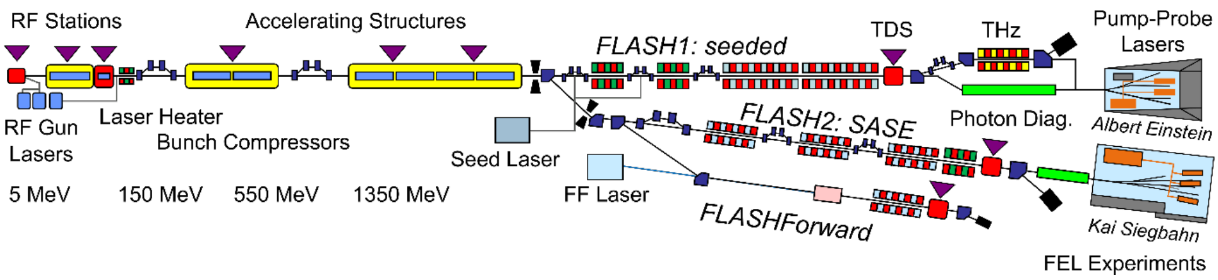

Figure 7: Schematic layout of the FLASH facility after completion of the FLASH2020+ project. The superconducting line is depicted on the left with its increased beam energy of 1.35 GeV max and the FEL beamlines to the right. For FLASH1, the HGHG and EEHG compatible seeding section followed by variable gap and polarization APPLE III type radiators as well as the THz generation with its bunch compression are illustrated. The branch below shows the upgraded FLASH2 SASE beamline optimized for short pulse generation is shown together with the FLASHForward beamline for beam-driven plasma acceleration studies. (Credit: DESY)

Conclusion and Outlook

The upgrade plans for both light sources at DESY, FLASH and PETRA III, and the close collaboration with interdisciplinary research institutions on site such as the CFEL, the CSSB, the CXNS, the CMWS, or EMBL, not to mention the nearby European XFEL facility, will open new avenues for a dynamic and unique research environment for photon science at DESY in the years to come. This development, however, will flourish only if the enormous amount of data generated at these facilities with the next generation of detectors can be handled, posing new challenges for scientific computing and data management. Initiatives in this direction have been started already. They will eventually shape and transform the way how interdisciplinary research at these facilities is performed in the future.

Acknowledgements

Funding for beamline P25 at PETRA III by the Freie und Hansestadt Hamburg and by the Bundesministerium für Bildung und Forschung (BMBF) in context of the Integriertes Technologie- und Gründerzentrum is greatly acknowledged.

The PETRA IV project team acknowledges funding of the technical design phase granted by the Behörde für Wissenschaft, Forschung, Gleichstellung und Bezirke (BWFGB) of the Freie und Hansestadt Hamburg under the contract BWFG/F 97236, as well as by the Bundesministerium für Bildung und Forschung (BMBF) under the contract DES21TDR.

The PETRA IV project team thank all researchers contributing to in the Scientific Instrument Proposals and all colleagues participating in the development and review of the PETRA IV beamline portfolio, in particular from EMBL, Hereon, KIT, MPG, India, and Sweden.

References

1. Abela *et al.*, Eur. Phys. J. Plus **138**, 355 (2023)
2. C. G. Schroer *et al.*, Phys. J. Plus **137**, 1312 (2022)
3. C. G. Schroer *et al.*, J. Synchrotron Rad. **25**, 1277 (2018)
4. C. G. Schroer *et al.*, PETRA IV: Upgrade of PETRA III to the Ultimate 3D X-ray Microscope — Conceptual Design Report (CDR), DESY (2019)
5. S. Antipov *et al.*, Phys. Rev. Acc. and Beams **24**, 111301 (2021)
6. Kroll *et al.*, Impact-Studie Synchrotronstrahlungsquelle PETRA III im Kontext des Forschungs- und Innovationsökosystems DESY, Fraunhofer-Institut für System- und Innovationsforschung ISI (2023)
7. Saldin *et al.*, Optics Communications **148**, 383 (1998)
8. M. Beye *et al.*, Eur. Phys. J. Plus **138**, 193 (2023)
9. W. Ackermann et al., Nature Photonics 1, 336 (2007)
10. B. Faatz *et al.*, Appl. Sci. **7**, 1114 (2017)
11. Sayed *et al.*, Chem. Rev. **122**, 10484 (2022)
12. Kroll *et al.*, Phys. Rev. Lett. **125**, 037404 (2020)
13. Maroju *et al.*, Nature **578**, 386 (2020)
14. Schneidmiller *et al.*, Photonics **10**, 653 (2023)